Numerical Study of Metal Oxide Schottky Type Solar Cells

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Abstract: Metal oxide (MO) semiconductors hold the promise for the development of high efficiency solar cells with low cost. Currently heterostructure type MO solar cells have been theoretically and experimentally studied, demonstrated their potential for applications. This paper highlights a numerical investigation on Schottky type MO solar cells using CuO as the absorption layer. It is shown that the doping concentration, absorption layer thickness, barrier height and back surface field have significant effects on the performance of the devices. Under the optimal structure and doping, the Schottky barrier solar cells, if can be fabricated with suitable techniques, can have a conversion efficiency up to 18.5%, comparable to MO heterojunction solar cells, but at a much simpler structure and lower cost. Some guidelines about the materials selection and structure design for MO Schottky barrier solar cells are summarized.

Keywords: solar cells, oxide semiconductor, Schottky diode, CuO, Cu_2O, CuO/Cu_2O heterostructure

1. Introduction

Photovoltaic devices are gaining increasing attention worldwide owing to energy shortage crisis, exhausting resources of natural fossil fuel and the corresponding greenhouse effect.

MO: Metal Oxide
BSF: Back Surface Field
MS: Metal/Semiconductor
MSV: Metal/Semiconductor/Voltage Enhancer
BH: Barrier Height
WAV: Window/Absorber/Voltage Enhancer

Currently, more than 90% of the photovoltaic market is taken by single-crystalline and poly-crystalline silicon solar cells that consume a large amount of energy to fabricate the materials and the devices. Furthermore the ever-increased prices for high grade-Si materials for microelectronics and photovoltaic industries prevent further reduction of the cost for Si-based solar cells, restricting the widespread deployment of PV panels for applications. Therefore, researchers and engineers around the world have been exploring new materials, advanced processes and novel device structures for better energy conversion efficiency and low cost to fulfil the rapidly increased demands for PV cells. III-V and II-VI compound semiconductors, $CuIn_xGa_{(1-x)}Se_2$ etc have been developed for manufacturing PV cells in homojunction or heterojunction configurations [1-3]. However, these types of cells suffer from problems of either shortage of natural reservation and toxicity of the elements such as Ga, As and Cd, or high manufacturing costs, severely limiting their use in PV cells. Organic solar cells are suffering from problems such as unstable operation and short life time due to rapid degradation and deterioration once exposed to natural environment, difficult for practical applications. Metal oxides such as TiO_2 and ZnO have been considered as promising candidates for PV applications, but have been utilized so far mostly to fabricate dye-sensitized solar cells that have shortages of low conversion efficiency, unstable properties and short lifespan. They are still at the early development stage, and are not suitable for large scale applications at the near future [4,5].

With the progress of technology, metal oxides such as Cu_2O and ZnO have also been considered for fabricating solar cells using traditional p/n junction or Schottky diode structures [6-8] as a large amount of theoretical studies have revealed that they potentially have good performances and are suitable for solar cells applications [9- 16]. However, practical MO heterojunction solar cells always show much poorer performance than the theoretical prediction. The highest conversion efficiency of ZnO/Cu_2O solar cell so far achieved is about 2%, which is much smaller than the theoretical efficiency of around 20 % [17]. The significant difference is believed to be mostly due to their high densities of interfacial states at the interface and defects within the oxide materials, partially due to un-optimized processes.

CuO has a narrow bandgap of 1.2eV, and is able to absorb much broader solar spectrum than Cu_2O; therefore it is a material with promise for low cost and high performance PV applications. It has not been studied thoroughly and used widely for electronic devices due to the difficulty in synthesizing the material in the past. With the improvement of deposition technology, high quality CuO layers with relatively good stoichiometry can now be obtained, which is indicative of its potential for the development of CuO-based solar cells and other electronic devices. We have conducted a theoretical investigation on CuO-based heterostructure solar cells with the transparent conducting oxides as the window layer [16], and showed that the theoretical performance of an ideal p/n junction type CuO solar cell can reach 17% for the two-layer TiO_2/CuO heterojunction and 28% when a back surface field (BSF) layer is added to the structure.

As an alternative to p/n junction solar cells, Metal/Semiconductor (MS) and its deviated structure of Metal/Insulator/Semiconductor (MIS) Schottky barrier solar cells are often used for fabricating solar cells owing to their simple structure and fabrication process, limited problems from the interface states for heterostructure introduced by semiconductor lattice mismatch etc, low temperature processing and reasonably good efficiencies [18- 25]. Various designs of Schottky diode structures and the effects of each semiconductor layer, metal electrode and electrode pattern on the performances have been systematically studied and explored for various material systems [26-28]. Silicon based 'OECO-MIS-1L' Schottky barrier type solar cells with conversion efficiency, $E_{FF,}$ ~15.7% were obtained in 1994, and the efficiency was further improved to 19.6% by using transparent conducting electrode and high quality thin SiN_x passivation layer, i.e. the so-called MIS structure [29,30].

MS and MIS type solar cells were initially considered for Cu_2O in 1980s [18,20]. A theoretical study showed an efficiency of 12% [31], though a much low practical efficiency of E_{FF}=1.8% was obtained due to imperfection of the materials and the M/S interface. Schottky barrier type diodes were also fabricated on some other MO semiconductors such as Ta_2O_5 and ZnO using Pt as the metal electrode though no PV effect was investigated [32,33].

Although Cu_2O is easy to be synthesized, its band gap is about 2.1eV [34] which corresponds to a wavelength of 590nm. As a result, the majority part of solar spectrum can not be absorbed by this material. On the other hand, CuO has an optimal band gap similar to Si, and the theoretical study has shown that CuO based heterojunction solar cells have much better performance than the Cu_2O solar cell with the same structure, having a comparative performance to Si cells. It is therefore important and interesting to study the CuO-based MS solar cell structures systematically, which may provide some novel structures with better performance, and guide the development of a new PV materials and PV cells for applications.

The present work will focus on the feasibility study of ideal CuO based MS and MSV solar cells based on computer simulation. The effects of various parameters of both the materials and structures will be studied and discussed.

2. Methodology

The computer simulation programme AMPS-1D was used for the study. It can be used to analyse dipole structures, such as homojunction and heterojunction solar cells. For Schottky diode type solar cell structures, the value of (E_C-E_F) at the metal/semiconductor interface can be set directly. The barrier height (BH) at both sides can be calculated as: BH= $\pm[\Phi_S-\chi_e-(E_C-E_F)]$, where Φ_S, χ_e, E_C, E_F are work function, electron affinity, conduction band energy level and Fermi energy level of the semiconductor; '+' is for the p-type semiconductor, while '-' is for the n-type semiconductor respectively. The value of (E_C-E_F) in the formula is closely related to metal work function. The other parameter settings for the simulation are listed in Table 1. The detailed material parameters and settings can also be found in ref. [16].

In this work, electrical properties of two structures of Schottky barrier cells are studied: MS and MSV structures (Fig.1). MS structure is a simple Schottky barrier diode with two metallic contacts on both sides of a semiconductor layer; MSV structure has an additional thin layer of a wide band gap material between the semiconductor layer and the back metallic contact of the MS structure. All the simulations were carried out under the assumptions of no bulk defects, under AM1.5 illumination at 300K. Only direct band-to-band recombination and surface recombination with a recommendation speed of 10^7cm/s at both the interfaces were considered.

The band diagrams of the MS and MSV structures are shown in Fig.2. The starting MS model is set as a p-type CuO layer of 1500nm with a narrow bandgap of 1.2eV and doping concentration of $1x10^{16}cm^{-3}$. Whilst the initial MSV model has an additional 100nm wide bandgap (2.1eV), p-type Cu_2O with a doping concentration of $1x10^{17}cm^{-3}$ at the backside. The front contacts are all Schottky type barriers, while the back contacts are all flat band or Ohmic if not specified.

3. Results and Discussion

3.1 Schottky barrier solar cell thickness

Figure 3 shows the effect of the CuO absorption layer thickness on the performance for the MS type device when the front contact was set to have a barrier height of BH= 0.8eV (E_C-E_F= 0.1eV) and a perfect Ohmic contact for the backside (BH= 0eV), with illumination from the front side and back side respectively.

For the front illuminated device, it is clear that the semiconductor layer thickness has a positive effect on all of the performance parameters: short circuit current (J_{SC}), open circuit voltage (V_{OC}) and fill factor (FF) and conversion efficiency (E_{FF}). The performance improves with increasing the thickness rapidly, and then slows down when the thickness increases beyond 500nm, and eventually saturates after 3000nm. CuO has a high optical absorption co-efficient, in the range of 10^4~10^5cm^{-1} [35,36]. A layer of a few micrometers in thickness is sufficient to absorb most of the incident light, so that increase in thickness beyond 3000nm does not improve the performance of the cell further. The fill-factor FF decreases slightly after about 3000nm due to the increased series resistance as the thickness increases.

The results show that an ideal Schottky barrier solar cell of 1500nm CuO layer can have a performance of J_{SC}~32.60mA/cm^2, V_{OC}~0.58V, FF~0.78 and E_{FF}~14.70%; for a 3000nm thickness CuO layer Schottky diode, it has E_{FF}~16.45%, with J_{SC}~35.48mA/cm^2, V_{OC}~0.59V and FF~0.79. Compared to TiO_2/CuO hetero-junction solar cell with 1500nm CuO layer [16], (J_{SC}~30.97mA/cm^2, V_{OC}~0.62V, FF~0.82 and E_{FF}~15.76%), it shows that the Schottky barrier cell has a slightly better J_{SC}, but a slightly decreased V_{OC} and FF. The smaller J_{SC} for the heterostructure cell is due to the optical reflection at semiconductor/semiconductor interface, which leads to some of the light reflected back to the wide band gap material.

Metals with suitable work function are essential to fabricate good Schottky barriers on semiconductors. These opaque contacts, even in grid form in the front, will reflect a significant amount of incident light, leading to a decreased efficiency. Solar cells with back illumination configuration are sometimes used for minimizing the light reflection by metal electrodes even for p-n junction type cells. The performance of the back illuminated MS cell is also shown in Fig.3 for comparison. Except the device with a layer thinner than 300nm, all the devices show a much worse performance compared to the front illuminated sample, and the efficiency decreases rapidly as the thickness increases. This can be explained as the highest carrier generation occurs in the depletion region for the front-illumination mode, but outside of the depletion region with no built-in electric field for the back-illumination mode. The optimal thickness of 300nm is approximately the depletion width of the diode at this doping level. If the cell thickness is set thinner than the depletion width for the back illuminated cell, all the electrons and holes generated can be separated effectively by the built-in electric field. If the cell is set thicker than the depletion width, most of the carriers generated will be in a region without built-in electric field, unable to contribute effectively to electricity generation, leading to decreased efficiency. For a Schottky barrier structure, the depletion region width is expressed as [37]:

$$W= (2\varepsilon_s V_{bi}/qN)^{0.5} \qquad (1)$$

Where ε_s is the semiconductor layer permittivity, V_{bi} is the built-in barrier; q is the electron charge and N is the doping concentration of the layer. The depletion width is in the order of several hundreds of nanometers for most of semiconductors if the doping levels and relative permittivities are similar. As a result, only those materials with a high optical absorption co-efficient ($>10^5 cm^{-1}$) will absorb sufficient light within the narrow depletion region for energy conversion. Cells fabricated with these materials will work most effectively under back-side illumination. For this purpose, the back Ohmic contacts should be made of transparent materials such as transparent conductive oxides (TCO).

3.2 Barrier height for both the contacts

Figure 4 shows the front contact Schottky barrier height effect on the cell performance. Under the assumptions used in the simulation, the highest possible front contact BH is about 0.9eV. It is obvious that the higher the barrier height, the better the performance; and all the parameters increase with the barrier height, though J_{SC} saturates much faster than the others. If the barrier height is lower than 0.5eV, the performance of the cell will be reduced by more than 50% compared to the best situation. With further decrease of BH, the cell will not work properly as it becomes an Ohmic-like device. As shown in eq.(1), the depletion width is strongly associated with the barrier height. Therefore, the BH, qV_{bi}, is one of the key parameters for Schottky type solar cells. Based on the work function theory, the barrier height is determined by the difference of the work function of a metal and affinity of the semiconductor. The barrier height for CuO can be varied from 0 to ~0.9eV by using various metals. However for practical Schottky diodes, the barrier height is strongly affected by crystal structure of the metals, contamination, perfection of semiconductor, interface states etc, and in most cases, it is smaller than that predicted by the work function theory. To improve the barrier height, a thin insulator layer is often inserted between semiconductor and metal to form the MIS structure which has been applied in Si-based solar cells [38].

For Schottky type diode, it is normally assumed to have a perfect Ohmic contact at the back. The imperfection of Ohmic contact will have a significant effect on the performance of the solar cells. As an indication, the effect of the back barrier height is studied with results shown in Fig.5. It was obtained by keeping BH=0.8eV for the front contact, while varying the back contact barrier from -0.3eV to 0.1eV. Figure 6 shows the cell band diagrams with different back contact BH settings. BH $\leq$ 0 represents a perfect Ohmic contact, and is the case for most Schottky diodes with heavy doping level as discussed later, while that with BH > 0 represents poor Ohmic contact, forming the so-called back-to-back Schottky diode.

It is obvious that the higher the back contact BH, the worse the cell performance though mainly the deteriorated V_{OC}. If the back contact is perfect Ohmic, such as BH=-0.3eV (E_C-E_F =E_G =1.2eV, Fig.6a), the cell can achieve E_{FF}~18.51% and V_{OC}~0.67V, significantly better than the case of BH=0eV, the flat band case, as shown in Fig.2a (E_{FF}~14.70% and V_{OC}~0.58V); However if the back contact is Schottky-like contact with BH>0, the performance would be deteriorated seriously as the barrier is not good for carrier transportation. An ideal Ohmic contact with BH$\leq$0 can improve the cell performance from 14.7% up to 18.5%, about 25% improvement compared to the flat band Ohmic contact.

3.3 Back surface field effect for MSV structure

By adding an additional thin layer of a wide bandgap material (e.g. Cu_2O, 2.1eV, 100nm) on the back side of the MS cell, we can obtain a MSV solar cell structure (Fig.1b and Fig.2b). Figure 7 shows the MSV cell performance as a function of CuO layer thickness. The performances for the front illuminated MS structure (shown in Fig.3) and the Window/Absorber/Voltage enhancer (WAV) heterostructure cell from ref. [16] are also plotted in the figure for comparison. Similar to the MSV structure, the WAV cell is based on TiO_2/CuO/Cu_2O, but TiO_2 is a n-type layer with a perfect Ohmic contact at the front. For the MSV structure, it further shows that the thicker the CuO layer, the better the performance is, unless the layer is too thick to reduce the fill factor significantly, and the corresponding E_{FF}. Generally, the MSV solar cell works better than the MS cells, especially on V_{OC} (0.1~0.2V better), mostly due to the BSF effect introduced by the wide bandgap semiconductor Cu_2O. The BSF effect introduced by heavily doping and by the conduction band offset shows a similar effect on the cell performance. The slightly improved J_{SC} is attributed to the back surface field effect. As shown in Fig. 8, the generation rate in Cu_2O V-layer is six orders of magnitude smaller than in the CuO layer, absorption in the V-layer does not make any contribution to Jsc.

The BSF layer, induced by either the band offset for heterostructures or the heavy doping on the back side of a homojunction structure, can block and reflect minority carriers diffusing to the back contact and make a contribution to V_{OC}. For a Schottky barrier solar cell, it is slightly different due to the nature of the majority carrier device. The BSF effect by the large conduction band offset (~0.9eV) at the CuO/Cu_2O interface can also block and reflect minority carriers diffusing to the back contact, but the effect is minor due to limited minority carrier concentration. A Cu_2O BSF layer can increase V_{OC} from 0.62V to 1.03V, an improvement of 70% for the WAV heterojunction cells, but can only increase V_{OC} from 0.58V to 0.69V, a 19% improvement for the corresponding Schottky barrier solar cells. The effect of the BSF layer on the performance is most effective when CuO layer is set to be thin, mostly attributed to the improved FF. This is slightly different from that for the WAV solar cell, which is mostly attributed to the enhanced V_{OC} [16]. Nevertheless, a V-layer should be introduced, if possible, to improve the Schottky type solar cells.

3.4 Comparison of CuO and Cu_2O Schottky barrier solar cells

Cu_2O has been intensively studied in various forms for the purpose of PV application. It is interesting to compare the performance with that of CuO diode. Figure 9 is a comparison of performances of ideal CuO and Cu_2O Schottky type (MS) solar cells as a function of layer thickness, simulated by the same AMPS-1D software. The results of Cu_2O MS solar cell are comparable to those obtained by Wang et al [31]. It is clear that the CuO MS solar cell has a much poorer V_{OC} (~ 1V smaller) and FF (~ 0.1 smaller) than those of Cu_2O MS cell, but much higher J_{SC}. This is mainly owing to the narrow bandgap of CuO (1.2eV) which is able to absorb much wider solar spectrum. Overall the CuO MS solar cell shows about 25%~30% better theoretical performance than the Cu_2O MS solar cell.

Cu_2O-based Schottky solar cells were previously fabricated with efficiency up to 2%. A maximum V_{OC} around 0.7~0.9V were always achieved regardless of the metals used, though much smaller than the theoretical value of about 1.5V. It is believed that there exists a copper-rich region at the interface caused by the reaction between copper-oxide layer and metallic contacts, leading to high Schottky barrier heights [39,40]. On the other hand, the barrier height for CuO Schottky diode is normally very small; an open voltage of a practical value of 0.7~0.9V and a theoretical value of 1.5V would be very attractive for the CuO solar cells. It is, therefore, natural to consider a thin Cu_2O as the surface layer for CuO Schottky solar cells to increase the V_{OC}, while maintaining the high J_{SC} of the CuO main layer. This situation was simulated with the results shown in Fig.10. Surprisingly a thin Cu_2O barrier layer (5nm) at the front side reduces the CuO cell efficiency significantly from E_{FF}~18.5% to less than 0.5%, though the Voc increases only slightly from ~0.67V to ~0.71V. J_{SC} of the structure with an ultra thin surface layer is reduced dramatically compared to the single layer CuO MS cell structure, because the carriers generated in the CuO layer are blocked by the barrier of the thin Cu_2O layer, and are unable to contribute to J_{SC} (Note: tunnelling through the barrier was not considered in the simulation. If the tunnelling is considered, a slight improvement is expected, especially for a thinner Cu_2O layer). This may have a serious implication for practical CuO Schottky solar cells as a copper-rich layer Cu_xO (x>1) may always exist at the surface of a CuO material, and may explain the low experimental efficiency normally obtained so far. Therefore, the surface of CuO should be carefully controlled to prevent CuO reduction to Cu_xO during fabrication.

4. Conclusions

High optical absorbing material CuO based MO Schottky barrier solar cells have been studied systematically. It is proven that these Schottky barrier solar cells, if can be fabricated with suitable techniques, can obtain conversion efficiency up to 18.5% with a 1.5μm thickness, comparable to MO heterojunction solar cells. Some guidance about Schottky barrier solar cell material selection and structure design are summarized below:

- Theoretically, the materials suitable for Schottky barrier solar cell is preferable to have small bandgap and very high optical absorption coefficient (> $10^5 cm^{-1}$). Therefore, sufficient incidence light can be absorbed by a very thin layer of material (about 1~2μm).
- It is desirable for the semiconductor layer to be lightly doped to have a wide depletion region in the device for better conversion. However, the maximum barrier height can be achieved is also limited by the low doping level.
- The front contact barrier height need to be larger than ~1/3 E_G of the semiconductor to achieve a reasonable performance. Therefore, p-type semiconductor is desired to posses a large electron affinity, and n-type semiconductor is desired to posses a small electron affinity to allow wide choice of metallic contacts which can form a satisfying barrier height with the semiconductor.
- It is desirable that the Schottky solar cell is front illuminated. For many p-type semiconductors, the suitable metal which can give a satisfying Schottky BH theoretically are opaque, which would reflect the incident light significantly. Back illuminated Schottky solar cells with sufficiently thin layer can be used to minimize light reflection and improve efficiency, but the efficiency is much lower than the front illuminated cells. In this case, the back Ohmic contact should be transparent conductive oxide.
- It is desirable to add a wide bandgap layer or a heavily doped layer on the back side of the device, to introduce a BSF effect which can improve cell efficiency by about 20%.
- Additional high barrier layer should be avoided at the front of CuO MS structure as it drastically reduces the performance.

Acknowledgement: The authors would like to acknowledge the financial support from the Technology Strategy Board under the grant number of TP11/LCE/6/I/AE142J and the Knowledge Centre for Materials Chemistry under the grant number of X00680PR.

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